RESUSPENSION OF SOIL PARTICLES FROM ROCKY FLATS CONTAINING PLUTONIUM PARTICULATES

G. Langer

Environmental Management
Air Quality & Chemical Tracking Division

EG&G Rocky Flats, Inc.
P. O. Box 464
Golden, CO 80402-0464

October 29, 1991

George H. Setlock
Reviewed for Classification/UCNI
By: George Setlock UNU
Date: October 29, 1991

EG&G ROCKY FLATS

EG&G Rocky Flats
Internal Publication
GHS-0070-91

Table of Contents

List of Figures

EXECUTIVE SUMMARY

This report presents an overview of research conducted at the Rocky Flats Plant (RFP) on the resuspension of soil particles from soil contaminated with plutonium (Pu) in the area called the "903 Field." This field is adjacent to and directly east of a former oil drum storage area which in 1969 was paved with asphalt and designated the "903 Pad." The 903 Field is a source of airborne Pu, due to wind erosion, and has been studied since 1970 for the resuspension rate of Pu particles. The following processes were considered:

- saltation (wind erosion of bare soil);
- wind resuspension of particles from grass blades;
- rain splash; and
- mechanical disturbances and grass fires.

Results indicate wind resuspension from bare soil seems to be minimal, while resuspension from grass appears to be the dominant process. Additionally, rain splash was also found to be a significant resuspension process. Over 90 percent of the resuspended Pu from the 903 Field is associated with soil and grass litter particles larger than 3 μm. The airborne radioactivity is roughly proportional to the mass of particles collected. Resuspension of respirable particles from the field is very limited; this respirable concentration at the field is about the same as that due to nuclear fallout in and around the Denver area. Maximum transport of the Pu extends to 1.5 km from the 903 Field. The release of Pu is parameterized by a resuspension factor of 5×10^{-11} m^{-1} and a resuspension rate of 2×10^{-12} sec^{-1}. The total resuspension is very low, estimated at ~200 μCi/yr. For a typical respirable particle concentration of 0.01 fCi /m^3* of Pu-239 near the 903 Field, the Pu collected was equivalent to one, 1-μm particle per month, using a sampling rate of 1.1 m^3/min.

* 1 fCi = 10^{-15} Ci

INTRODUCTION

Concern over public health in regards to the 903 Field, located adjacent to a former outdoor drum storage area for waste oil, is recognized by RFP. The plant has been monitoring this area since the first oil drum leaks were discovered 30 years ago. The waste oil in these drums contained residue particles less than 3 µm in size of Pu from machining operations. Removal of the drums began in 1967 and the area was partially remediated and subsequently covered with an asphalt pad in 1969. During this period and continuing through the present, air at the 903 Field adjacent to the pad and in various locations around the Denver area (Figure 1) is continuously monitored for airborne Pu/soil particles by a network of surveillance air samplers. In the discussions that follow, it should be kept in mind that the Pu is attached to host soil particles that range in size from a few micrometers to millimeters. This is due to the nature of the original contamination process.

At no time since the completion in 1971 of the drum storage clean-up has Pu concentration exceeded the DOE "Derived Concentration Guide" of 20 fCi/m^3, either at the source area or in the surrounding community. In fact, it can be noted the concentration of respirable Pu particles at the 903 Field is near background levels found in the Front Range area of Colorado. Taking into account all significant pathways of human Pu intake, exposure to Pu at the 903 Field is well below EPA proposed guidelines (EP78, p221)*. The average total radioactivity concentration of airborne Pu at the plant boundary is 0.05 fCi/m^3.

The 903 Pad and Field are scheduled for further investigation and remediation in the future. The alpha radioactivity in the 903 Field soil is much less than EPA proposed guideline levels. The alpha radioactivity from RFP waste that has entered the environment amounts to a few curies, while waste tailings (uranium and thorium) from mining activity amount to a few thousand curies in an area in downtown Denver (KA84, p130). A synopsis is provided in this report of RFP research on the resuspension of Pu particles from the 903 Field. This research included:

- extent and radioactivity characterization of the source area;
- consideration of all feasible processes of resuspension; and
- investigation of the subsequent transport of the airborne particles according to their size and radioactivity category.

---

* The last part of the literature citation, following the "p", indicates the page number at which the information will be found.

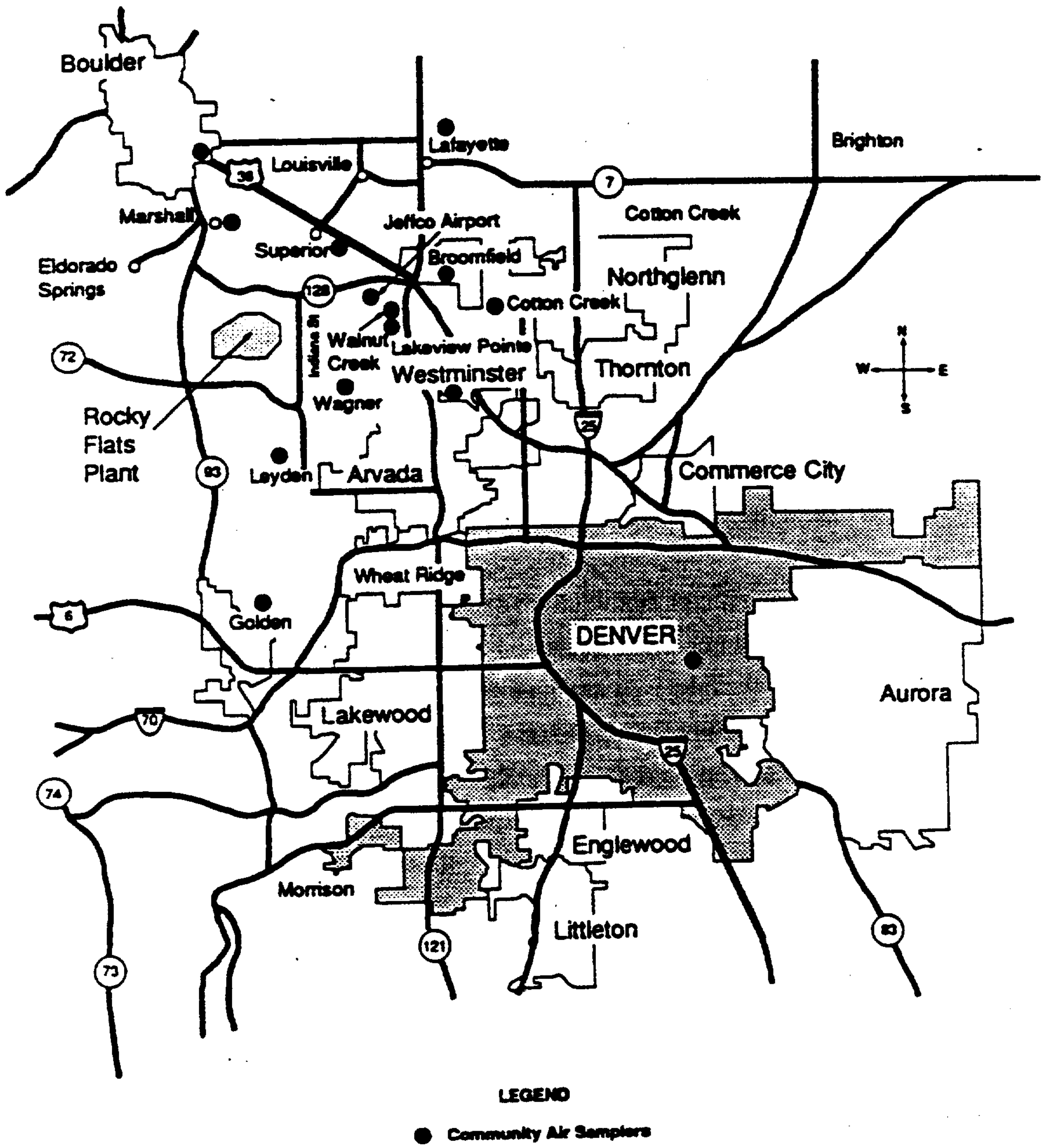


Figure 1. Area Map and Locations of RFP Community Samplers

SOURCE AREA AND SOURCE ACTIVITY

The area now known as the 903 Pad, after removal and off-site shipment of oil-covered surface rocks, was covered with gravel and then asphalt in 1969 to immobilize Pu-contaminated soil particles (Figure 2). However, during site preparation for the asphalting, occasional high winds swept across the uncovered area. Some dust was generated and much of it settled a short distance to the east of the site towards the plant site security fence. This area near the security fence is designated the 903 Field and has been covered with off-site topsoil. Vegetation has also been re-established. During the stabilization process fugitive Pu particles in the sub-surface soil were mixed into approximately 20 cm of new topsoil. This allows the possibility that some Pu particles could be resuspended, due to water or wind erosion. Therefore, access to the area is restricted and the ambient air in this area is continuously sampled by RFP for plutonium.

It should be noted that mine tailings from radium extraction, as found in some locations within the city of Denver, and mine tailings used in Grand Junction for home construction pose greater health hazards than the Pu contamination present in the 903 Field. Mine tailings release radon gas, which is difficult to immobilize, and the radon decay products become attached to respirable dust particles. The RFP Pu particles are relatively immobile and require extensive force to become airborne, which results mostly in particles too large (> 10-μm) to be inhaled (HA80a, p216). Once the small (<3-μm) Pu particles in the waste oil were immobilized by attachment to soil particles they became very difficult to separate, due to interatomic, attractive surface forces.

Figure 2 also shows the distribution of Pu in soil at and near the plant as determined by the Atomic Energy Commission (AEC) Health and Safety Laboratory (HASL) (RF3115, p14). The amount of Pu that leaked from the drums throughout their existence in the 903 Pad area was estimated in 1971 as 6.1 Ci or 86 g (SE71, p6; EI80, p2-73) based on the amount of oil leaked and Pu content of the oil. Nearly 4.0 Ci are now immobilized under the asphalt pad. A detailed ground gamma survey (RF3689, p18) for the Am-241 associated with the Pu indicated that approximately a total of 1.2 Ci of Pu exists in the 903 Field area west of the perimeter fence and bounded to the west by the 903 Pad. This survey was made after the removal in 1978 of soil containing an estimated 0.5 Ci of Pu-239, along the hill crest on the southern edge of the 903 Field. Another 0.67 Ci of Pu exists on the east side of the security fence in a small, localized area. Figures 3 and 4 illustrate this ground Am-241 gamma survey. These regions of high Pu soil concentrations are considered the source area for chronic release of Pu from RFP and this soil will likely be removed in the future once an acceptable regulatory framework has been worked out.

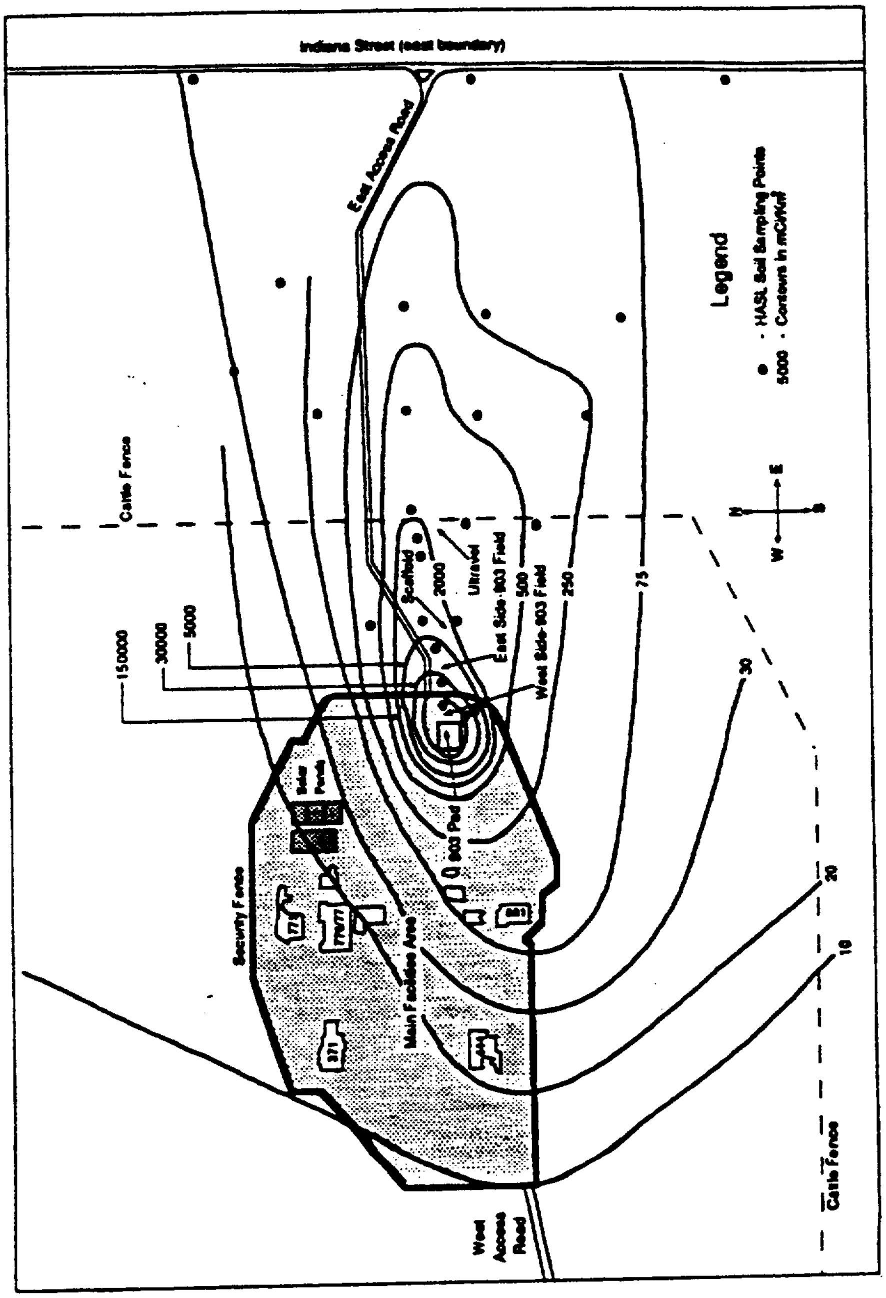


Figure 2. Plutonium-239 Deposition Contours in Millicuries Per Square Kilometer, According to HASL

Enlargement of 903 Pad Area

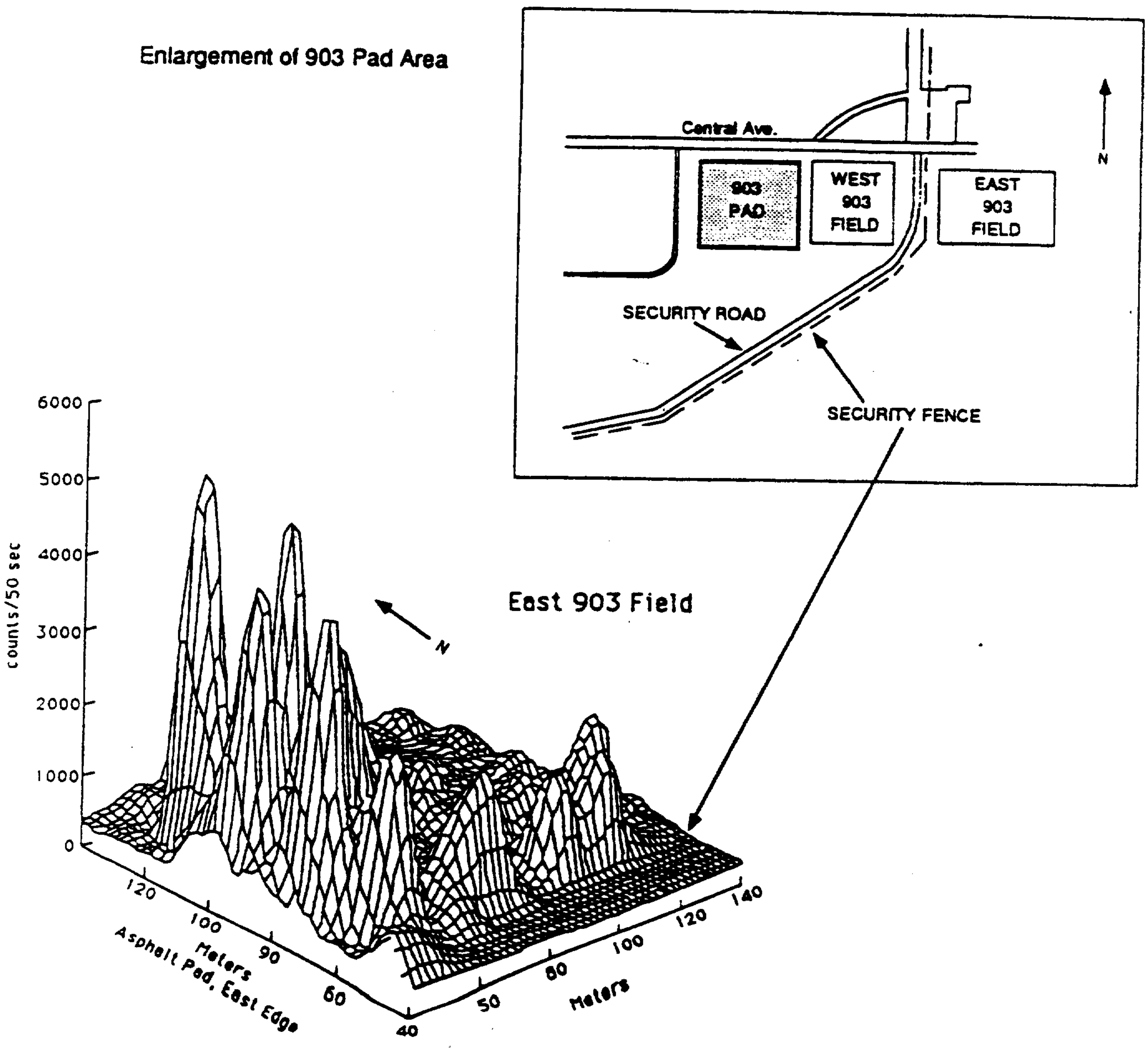


Figure 3. Three-deminsional Plot of Gamma Activity from Survey of West Side of 903 Field

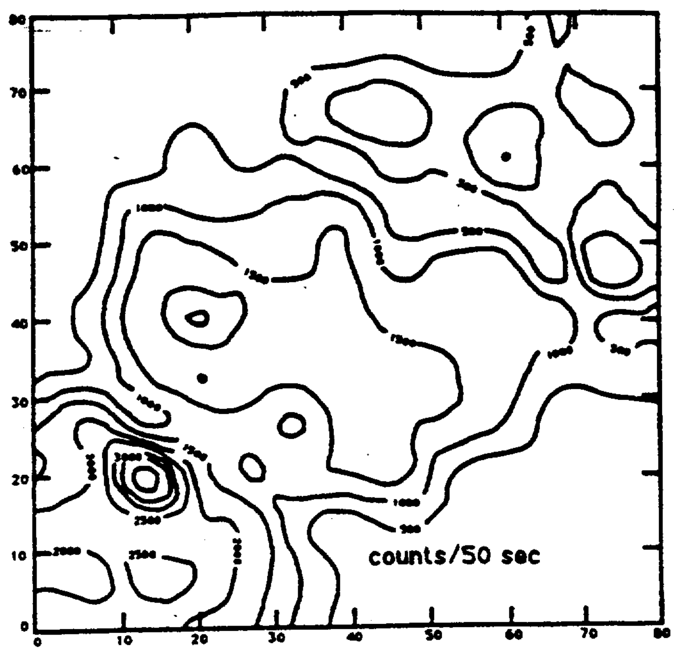


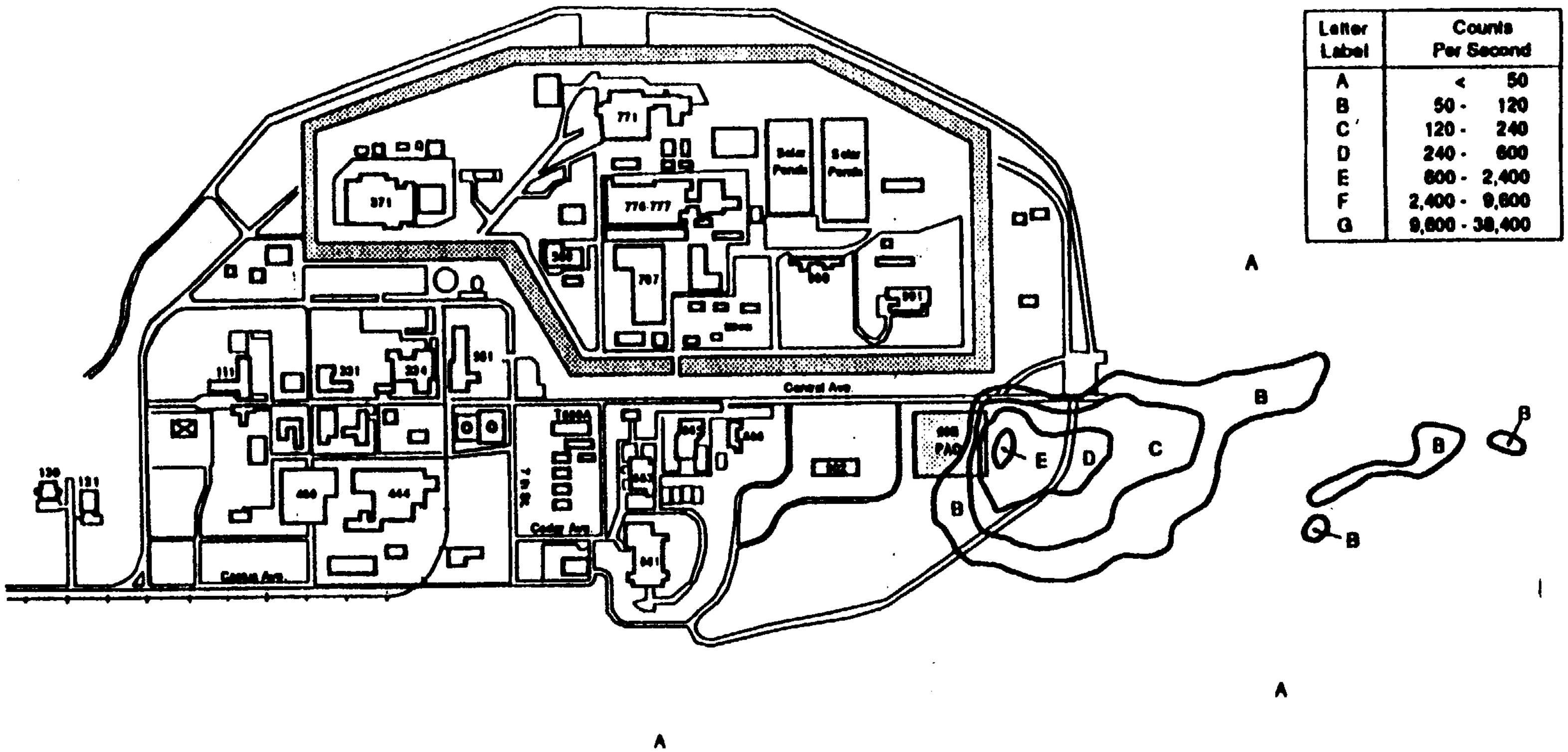
Figure 4. Contour Plot of Gamma Activity from Survey of East Side of 903 Field

P. W. Krey, in "Remote Plutonium Contamination and Total Inventories from Rocky Flats" (KR76, p214) has estimated, based on limited samples, that in addition to the Pu in the 903 Field there is another 3.4 Ci spread out over a wider region. This region, with near fallout Pu levels, extends east and southeast from the security fence. This would indicate that, based on a material balance, more than 9 Ci Pu-239 leaked from the drums instead of the 6.1 Ci estimated in 1971 and a total of about 5 Ci were resuspended from the drum storage area. Once past Indiana Street, the current eastern boundary of the plant, only a small percentage of the Pu that has been found is of RFP origin, as explained below. The rest is from residual fallout from past global atmospheric weapons testing. An isotopic ratio (Pu-240/Pu-239) analysis was used by Krey to distinguish the RFP contribution from atmospheric fallout to one-tenth of fallout levels. The RFP contribution in the region east of Indiana Street is approximately 0.1 Ci total, according to Krey.

Another possible source of RFP Pu in the environment is from safety shots, i.e., non-fission detonations, at the Nevada Test Site (NTS). Plume deposition from these shots has been tracked as far east as Grand Junction, Colorado, where it contributes 0.6 mCi/km^2 to the Pu in soil (BE83, p23). Global fallout away from Nevada is of this order. The dispersion of NTS Pu has not been investigated east of Grand Junction.

Also of interest are the periodic aerial gamma surveys of the plant and surrounding land by helicopter using an array of sodium iodide crystal detectors (BO82; BO90). Figure 5 presents the 1989 survey contours for Am-241 (BO90). The 1981 aerial survey data show similar results (BO82). In 1989 the sensitivity of the aerial instrumentation was improved, making it possible to detect Am-241 beyond the cattle fence which marked the plant's original boundary (e.g., to the 250-mCi/km^2 contour shown in Figure 2). Also, the 1989 survey extended beyond the confines of the RFP buffer zone to obtain a broader picture of background radioactivity. As far as total radioactivity is concerned, which includes natural radioactive sources, the hottest localized spot was an old mine shaft near Leyden, several miles from RFP. The major contributor to this radioactivity was Bi-214 from radon gas (BO90, p11). To detect the above background radioactivity at RFP the survey has to be specific for Am-241.

The airborne surveys showed that Pu radioactivity has not migrated significantly beyond the original source area after the 1969 asphalt stabilization of the 903 drum storage area (BO90, p25). The 1989 study also included selected ground sampling points for Am-241 measurements (Figure 6). A portable gamma spectrometer was used for this purpose, to be followed by radio-chemical analysis for Pu from soil samples from the same location.



Letter Label	Counts Per Second
A	< 50
B	50 - 120
C	120 - 240
D	240 - 600
E	600 - 2,400
F	2,400 - 9,600
G	9,600 - 38,400

Figure 5. Aerial Gamma Survey - 1989

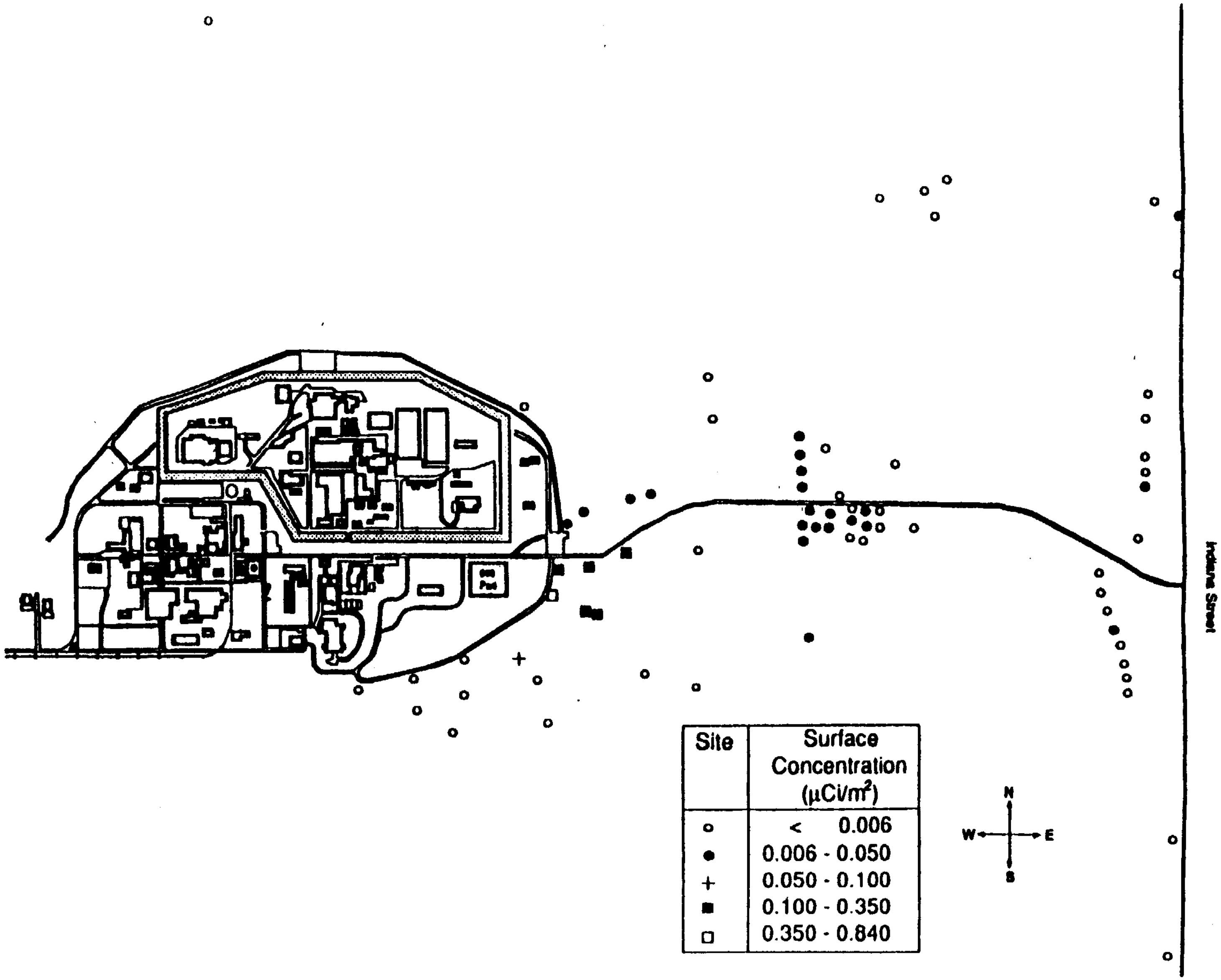


Figure 6. Gamma Ground Survey Sites for Am-241

PHYSICS OF SOIL RESUSPENSION

The potential for chronic release of airborne Pu particles from RFP is limited to soil resuspension from the 903 Field. Building releases from High Efficiency Particulate Air (HEPA) filters are small, i.e., a total of 5 µCi/yr in 1989 (RFENV-89, p23) vs. 200 µCi/yr from the 903 Field (see section on resuspension factors). We know from air monitoring that Pu particles do become airborne from soil, but the physics of the process need to be understood for dispersion modeling and remedial action purposes. Soil particles are traditionally thought to be resuspended by wind forces acting on bare soil surfaces, but studies upon which this view is based have been limited to plowed fields and desert areas. The 903 Field has only small bare soil areas between the clumps of bunch grass, but the original premise of the author and others was that the Pu particles originate from the bare soil between the clumps of grass.

Saltation

Initial resuspension studies were directed at the classical resuspension process of saltation (HA80a, p213). That is, the wind propels millimeter sized particles that protrude beyond what is called the boundary layer (the immobile or stagnant air layer approximately 1 mm in depth, adjacent to the land surface) in a series of small hops. On impact these large particles knock loose smaller particles, in a manner similar to sand blasting. The smaller particles then become entrained into the main air streams by turbulence to heights over 6 m at a distance of less than 30 m (HA80a, p222).

It remained for RFP to demonstrate if saltation could occur from small areas devoid of vegetation, because this soil surface condition has received little attention from soil scientists. Direct visual observations initially indicated that even during wind storms reaching over 100 mph, no visible quantities of dust were released (e.g., puffs of dust from bare areas). Operation of a modified Bagnold Catcher for several week-long runs, including operation during wind storms, provided no weighable dust fractions. The Bagnold Catcher (GI74) is the classical device for measuring wind erosion of soil. Nevertheless, more sensitive techniques were developed to verify if saltation processes occur on a small scale.

One technique developed by RFP was the application of a ribbon-like laser beam grazing the soil surface, to detect impacting large particles and bursts of numerous small particles

(RF3197, p8). This was done at night using time exposure photography. No particles were observed until winds exceeded 35 mph, but even then resuspension was tenuous and no clear evidence for saltation was found by this approach (RF3325, p3).

The second methodology, shown in Figure 7, involved the placement of an acoustic particle detector underground, facing a 2.5-cm opening in a soil surface area devoid of vegetation (RF3115, p11). There was no net airflow into the opening, but the objective was to detect particles over 50 µm aerodynamic equivalent diameter (AED) that were resuspended by the wind and then fell back to the ground. As the particles fell back, some dropped into the intake of the acoustic particle counter. This system could operate continuously and unattended for several days. Again, no convincing case could be made for saltation, even in high winds (RF3115, p13; RF3197, p7). Evidently, the soil is too crusty for wind erosion (HA80a, p224), except for a few small areas (RF3115, p11). Also, many of the smaller, bare areas are too deep in the grass canopy to experience the full force of the wind. Only deliberate disturbance of the ground with a stick was observed to release short bursts of particles.

The above results may seem to contradict a study by Sehmel, who developed a Pu resuspension model for the 903 Field based on the saltation process (SE72). However, this model was based on data collected by RFP from July 1970 through January 1971. In March 1970 the 903 Field was disturbed by a major ditch construction project near the west side of the perimeter fence. It took nine months for the effects of this operation to disappear, i.e., for the loose surface soil to become crusty again and for introduced grasses to grow.

Next, resuspension studies were carried out under controlled conditions using the small wind tunnel shown in Figure 8 (RF3197, p5). The objective was to observe resuspension from bare ground as well as from grassy areas. Testing of bare spots showed very little release until extreme wind velocities (e.g., equivalent to 150 mph at 10 m or about 30 mph near ground levels) were applied or the soil had previously been disturbed. But even the latter, "fresh" surface was quickly exhausted of particles (RF3689, p36). This wind tunnel, at high flow, proved to be a useful approach to soil surface sampling for Pu particles and was extensively used for this purpose (RF3689, p23).

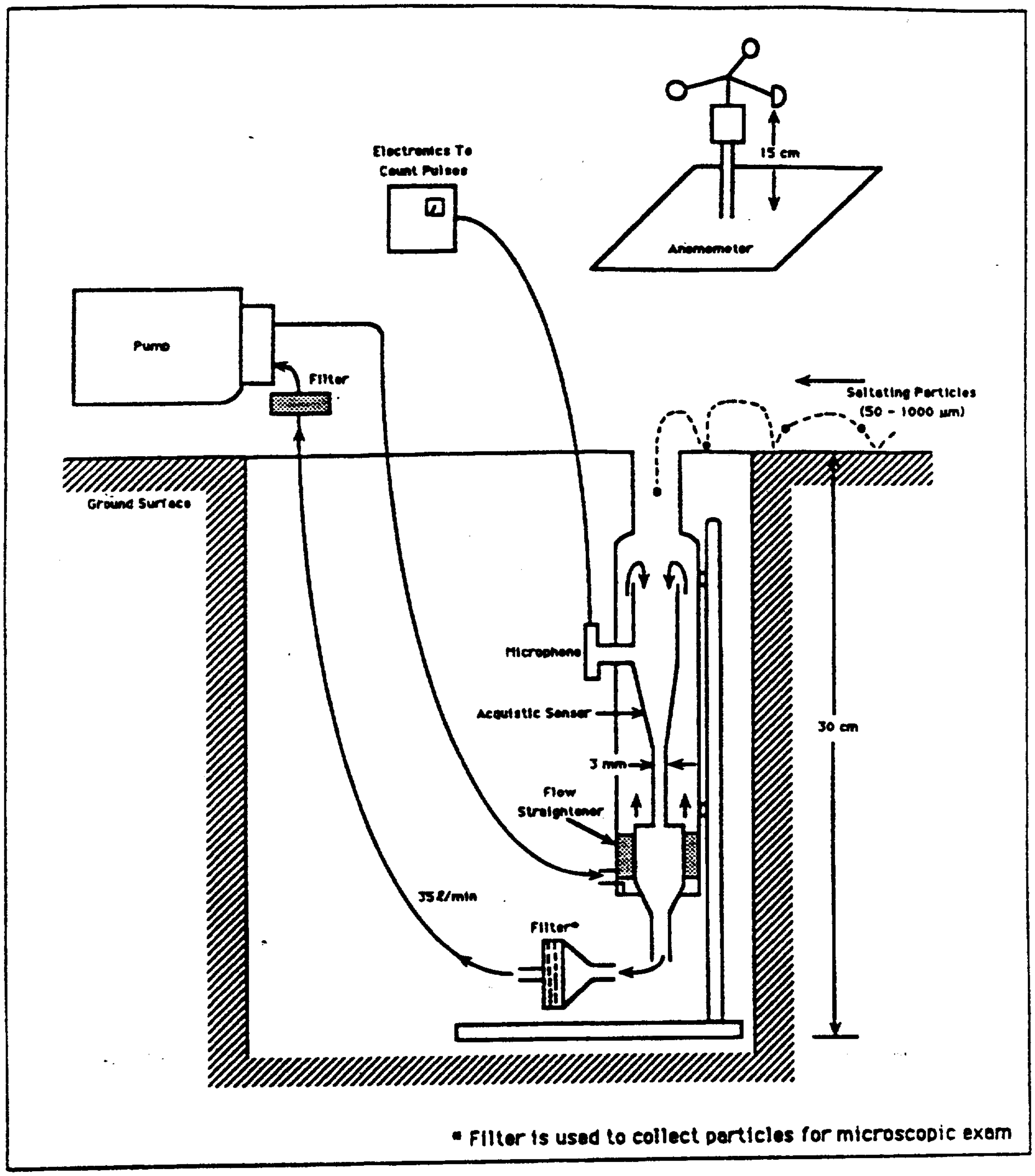


Figure 7. Acoustic Particle Counter for Saltating Particles

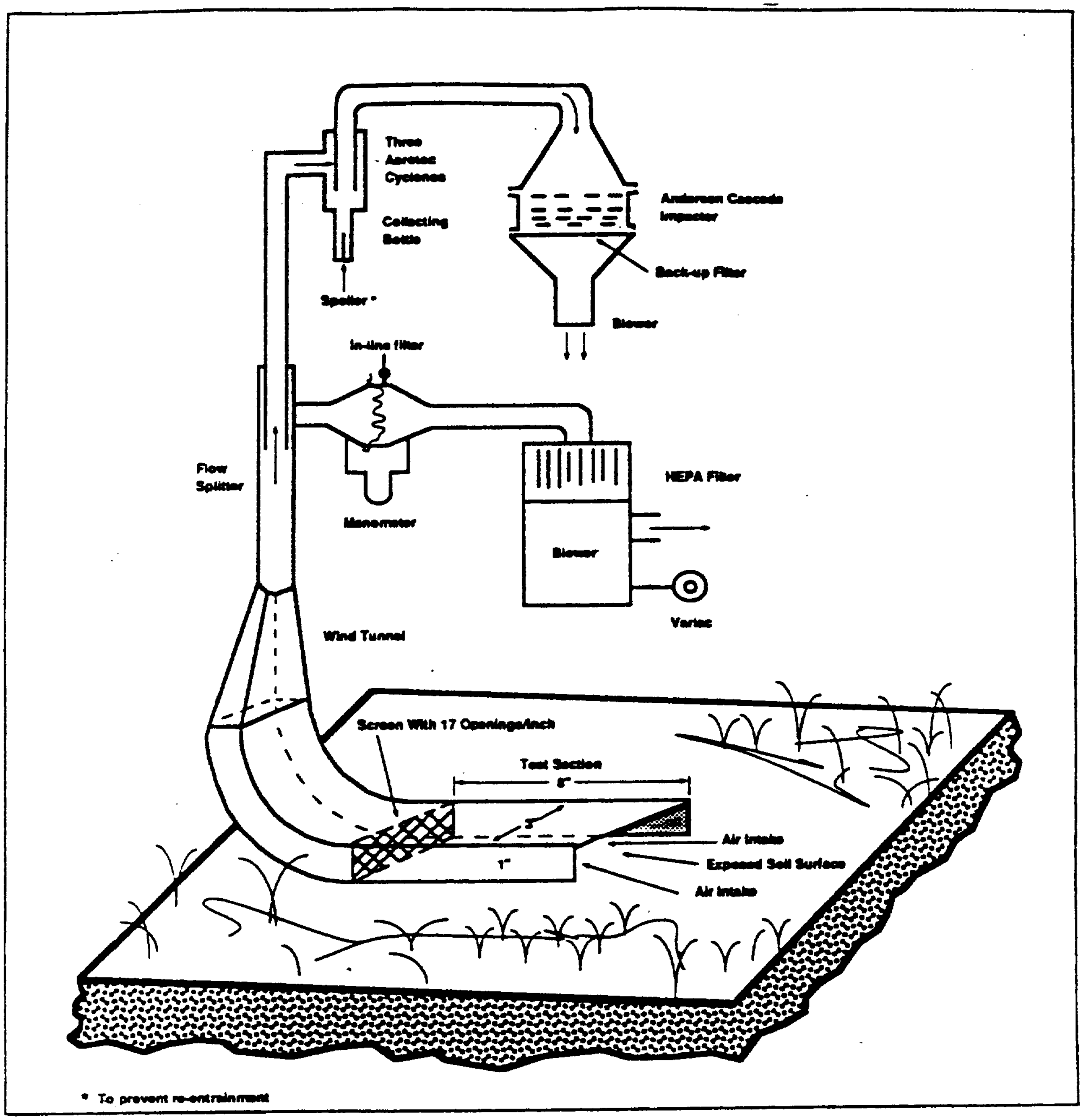


Figure 8. Portable Wind Tunnel for Resuspendable Dust

The wind tunnel was also operated on patches of grass, although it was not designed for this purpose. The height of the wind tunnel test section was less than the height of the grass. For proper testing of grass covered areas a considerably larger wind tunnel is required, such as the Gillette device (GI78). Nevertheless, even at low velocities (e.g., wind speeds equivalent to 20 mph at 10 m) the small wind tunnel detected small but significant amounts of resuspended Pu-carrying particles. This was considered important, since most of the ground in the 903 Field is grass covered. Much of the resuspended material was over 10 μm AED and was organic (i.e., grass litter) as shown by ashing the samples. The organic content was about 40 percent on the average (RF4036, p23).

Resuspension from Grass Blades

As a result of the aforementioned wind tunnel studies, attention was now focused on the details of resuspension from grass covered areas. It was not certain whether Pu resides on the grass blades and then becomes resuspended or if it originates from the grass litter on the soil surface and, as the grass decays, is then resuspended as part of decomposed grass particulates. Research was conducted to determine if both processes could be taking place.

First, it was verified quantitatively that Pu does reside on grass and grass litter (RF3914, p6). This was done by clipping the grass at successively lower levels and measuring each level for Pu distribution in relation to ground height. The Pu concentration in the underlying grass litter was also measured. Pu radioactivity was distributed fairly uniformly in relation to height for a total of 1.1×10^4 pCi for grass grown on a square meter of soil with a radioactivity of 2200 pCi Pu-239/gm of soil. The grass litter held 510 pCi Pu-239/gm of litter. Therefore, live grass must be considered a major source of Pu particles for resuspension in addition to the underlying grass litter. It should be noted that the litter is not readily accessible for resuspension because it is shielded from wind by the live grass.

The question of how the Pu becomes attached to the grass is of interest. Plant uptake of Pu is not a factor, because we are dealing with insoluble Pu particles (AR82, p33). This leaves wind driven soil particles from bare soil areas and rain splash as the source of Pu. The latter process is well documented for transferring Pu to vegetation to heights up to 30 cm (DR84, p183). Finally, the growth process of the grass as it rises through the soil surface was considered as a means of transferring Pu to the blades, but the grass blades start from the stem of the plant after it has risen out of the ground. Therefore, no direct transfer of soil particles to the blades takes place.

Information was now required on the capacity of grass blades to hold soil particles and on potential Pu resuspension mechanisms from the grass. Therefore, the surface of grass blades was studied with a scanning electron microscope. Most grasses at RFP have blades whose surface is covered with fine fibers that act like a filter matrix which intercepts considerable amounts of dust (RF4036, p15). C. Gutfinger reports that fibrous elements extending from a surface into the viscous boundary layer enhance deposition by a factor of 10 to 1000 (GU85, p3). The microscopy showed that the blades were heavily loaded with soil particles. Conversely, dust particles should be released when the grass fibers decay and fall off and when the blades flex due to wind. Such behavior was verified with the wind tunnel tests described below.

In a small laboratory wind tunnel (RF4036, p23) samples of grass were placed in the test section and exposed to air velocities that would be found at grass level due to winds of 5 to 20 mph at a height of 10 m above ground. From a 5-cm blade of grass about 200 particles were released in the 0.2 to 12-µm range, as verified with an optical particle counter and membrane filter samples. Most pertinent were tests where the blades were mechanically flexed, which simulated wind motion. Here, the release from the blades of particles greater than (>)10 µm was dominant, with a median diameter of 20 µm and a maximum of 40 µm.

To verify the above results in the field, a simple test with the soil resuspension wind tunnel was made in summer with the ground soaked with water but the grass dry (RF3914, p8). The object of this test was to demonstrate how much Pu is resuspended from grass blades alone. The blades are much more exposed to the wind than the ground but may hold dust more tenaciously. The resuspension rate was about one-sixtieth of that for a similar dry area at a wind speed equivalent to 80 mph at 10 m. At 20 mph it was one-fortieth less than at 80 mph. But these resuspension rates could still account for most of the radioactivity observed by the air samplers, since 95 percent of the field is covered with grass. These data have to be interpreted with some caution, because as pointed out above, the wind tunnel was not of an optimal design for studying resuspension from grass.

This test confirmed that the release of radioactive particles from grass blades alone is important, if not dominant. Additional radioactivity exists on dead grass litter on the ground between the standing grass, but this material is not readily available for resuspension because it is protected from the wind by the stands of grass.

Rain Splash

To determine the amount of Pu resuspension when the soil is completely saturated during long periods of rainfall, such as that encountered in spring (RF3914, p9) a series of tests were conducted. For this limited test series the airborne Pu concentration during rainfall did not differ significantly from that during dry periods. Rain splash was therefore studied as a means of releasing Pu particles into the air. First, a laboratory wind tunnel was set up to simulate single raindrops splashing on soil under controlled conditions. Provision was made to count resuspended soil particles by concentration and size with an optical particle counter. This experiment showed that soil particles do become aerosolized by rain splash, if a thin water layer exists on the soil surface (RF4036, p30).

Evidently, these airborne soil particles are the residuals that remain upon the evaporation of the hundreds of small satellite droplets that form along with big splash drops (GR73, p57). The satellite droplets are small enough to be carried by air currents.

This resuspension process was also field tested. A small plastic tent was built over a patch of Pu-contaminated bare soil at the 903 Field (Figure 9). The tent was necessary to prevent airborne Pu particles from drifting into the test area from the surrounding area. Nuclear track foils were placed on the resuspended residue particles collected from the splashes to verify the presence of Pu particles. The tests showed that soil particles containing Pu did become airborne due to drop impact (RF4036, p30). A thousand 5-mm rain drops resuspended 5 pCi into the air from soil with a surface radioactivity concentration of 2500 pCi/g. About 500 million raindrops may fall on an area of one square meter annually. To complete this analysis, the washout of resuspended soil particles by rain drops should be accounted for in a real situation (GR73, p121). The washout effect was not present in the aforementioned single drop experiment. The washout effect decreases the airborne Pu concentration as falling raindrops sweep out dust particles in the air.

Grass Fires

Another potential source of resuspended particles is the ash from grass fires. So far no fires have taken place in the 903 Field, but RFP has conducted tests to simulate such an

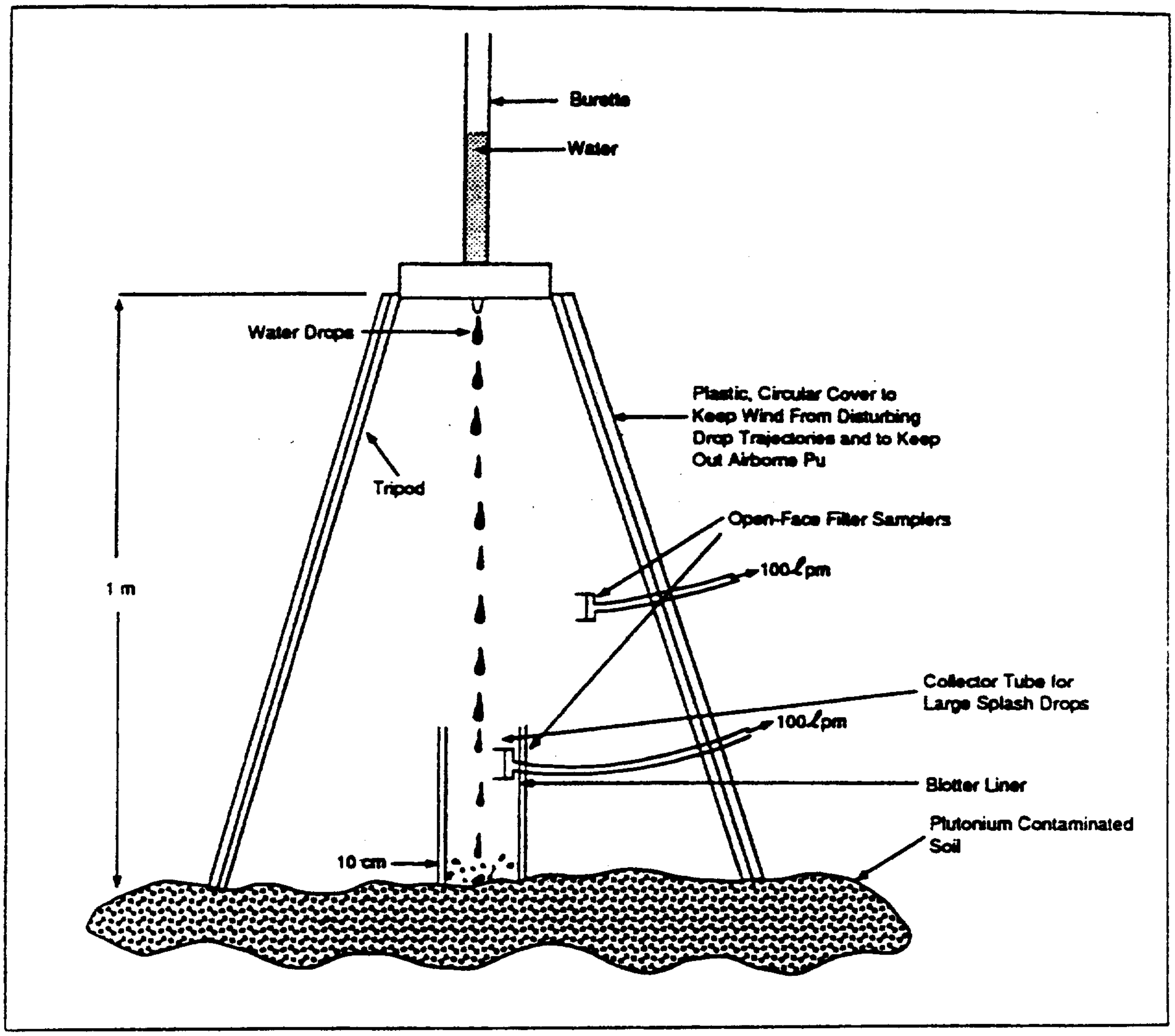


Figure 9. Simulation of the Resuspension of Plutonium-contaminated Soil Particles by Rain Splash

occurrence. Fire was set in a wind tunnel to 180 gm of grass collected from an area of soil 1 m^2 in size. The grass had a radioactivity concentration of 8.1 pCi Pu-239/g of air-dried grass. Smoke from this test fire had a total radioactivity of 34 pCi of Pu-239 or 17 pCi/g (LA86, p91). However, placed in perspective, consider that the annual limit on intake for a member of the public for Pu-239 is 170 pCi (DO90). If the whole field (0.02 km^2) were to burn, a person remaining in the plume would inhale a small fraction of this limit.